

Hierarchical Theory of Condensed Matter: Long relaxation, macroscopic oscillations and the effects of magnetic field

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[1]. A. Kaivarainen. Book: Hierarchic Concept of Matter and Field. Water, biosystems and elementary particles. New York, NY, 1995, ISBN 0-9642557-0-7

[2]. A. Kaivarainen. New Hierarchic Theory of Matter General for Liquids and Solids: dynamics, thermodynamics and mesoscopic structure of water and ice (see URL: <http://www.karelia.ru/~alexk> [see New articles]).

[3]. Hierarchic Concept of Condensed Matter and its Interaction with Light: New Theories of Light Refraction, Brillouin Scattering and Mössbauer effect (<http://www.karelia.ru/~alexk> [see New articles]).

[4]. A. Kaivarainen. Hierarchic Concept of Condensed Matter : Interrelation between mesoscopic and macroscopic properties (see URL: <http://www.karelia.ru/~alexk> [see New articles]).

See also set of previous articles at Los-Alamos archives:
http://arXiv.org/find/physics/1/au:+Kaivarainen_A/0/1/0/all/0/1

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Summary of new Hierarchic theory, general for liquids and solids

A basically new hierarchic quantitative theory, general for solids and liquids, has been developed.

It is assumed, that unharmonic oscillations of particles in any condensed matter lead to emergence of three-dimensional (3D) superposition of standing de Broglie waves of molecules, electromagnetic and acoustic waves. Consequently, any condensed matter could be considered as a gas of 3D standing waves of corresponding nature. Our approach unifies and develops strongly the Einstein's and Debye's models.

Collective excitations, like 3D standing de Broglie waves of molecules, representing at certain conditions the mesoscopic molecular Bose condensate, were analyzed, as a background of hierarchic model of condensed matter.

The most probable de Broglie wave (wave B) length is determined by the ratio of Plank constant to the most probable impulse of molecules, or by ratio of its most probable phase velocity to frequency. The waves B are related to molecular translations (*tr*) and librations (*lb*).

As the quantum dynamics of condensed matter does not follow in general case the classical Maxwell-Boltzmann distribution, the real most probable de Broglie wave length can exceed the classical thermal de Broglie wave length and the distance between centers of molecules many times.

This makes possible the atomic and molecular Bose condensation in solids and liquids at temperatures, below boiling point. It is one of the most important results of new theory, which we have confirmed by computer simulations on examples of water and ice.

Four strongly interrelated new types of quasiparticles (collective excitations) were introduced in our hierarchic model:

1. *Effectons* (*tr* and *lb*), existing in "acoustic" (a) and "optic" (b) states represent the coherent clusters in general case;
2. *Convertons*, corresponding to interconversions between *tr* and *lb* types of the effectons (flickering clusters);
3. *Transitons* are the intermediate $[a \rightleftharpoons b]$ transition states of the *tr* and *lb* effectons;

4. *Deformons* are the 3D superposition of IR electromagnetic or acoustic waves, activated by *transitons* and *convertons*.

Primary effectons (*tr* and *lb*) are formed by 3D superposition of the **most probable standing de Broglie waves** of the oscillating ions, atoms or molecules. The volume of effectons (*tr* and *lb*) may contain from less than one, to tens and even thousands of molecules. The first condition means validity of **classical** approximation in description of the subsystems of the effectons. The second one points to **quantum properties of coherent clusters due to mesoscopic molecular Bose condensation**.

The liquids are semiclassical systems because their primary (*tr*) effectons contain less than one molecule and primary (*lb*) effectons - more than one molecule. *The solids are quantum systems totally because both kind of their primary effectons (tr and lb) are molecular Bose condensates. These consequences of our theory are confirmed by computer calculations.*

The 1st order [*gas* \rightarrow *liquid*] transition is accompanied by strong decreasing of rotational (librational) degrees of freedom due to emergence of primary (*lb*) effectons and [*liquid* \rightarrow *solid*] transition - by decreasing of translational degrees of freedom due to Bose-condensation of primary (*tr*) effectons.

In the general case the effecton can be approximated by parallelepiped with edges corresponding to de Broglie waves length in three selected directions (1, 2, 3), related to the symmetry of the molecular dynamics. In the case of isotropic molecular motion the effectons' shape may be approximated by cube.

The edge-length of primary effectons (*tr* and *lb*) can be considered as the "parameter of order".

The in-phase oscillations of molecules in the effectons correspond to the effecton's (a) - *acoustic* state and the counterphase oscillations correspond to their (b) - *optic* state. States (a) and (b) of the effectons differ in potential energy only, however, their kinetic energies, impulses and spatial dimensions - are the same. The *b*-state of the effectons has a common feature with **Frölich's polar mode**.

The (*a* \rightarrow *b*) or (*b* \rightarrow *a*) transition states of the primary effectons (*tr* and *lb*), defined as primary transitons, are accompanied by a change in molecule polarizability and dipole moment without density fluctuations. At this case they lead to absorption or radiation of IR photons, respectively.

Superposition (interception) of three internal standing IR photons of different directions (1,2,3) - forms primary electromagnetic deformons (*tr* and *lb*).

On the other hand, the [*lb* \rightleftharpoons *tr*] *convertons* and *secondary transitons* are accompanied by the density fluctuations, leading to *absorption or radiation of phonons*.

*Superposition resulting from interception of standing phonons in three directions (1,2,3), forms secondary acoustic deformons (*tr* and *lb*).*

Correlated collective excitations of primary and secondary effectons and deformons (tr and lb), localized in the volume of primary *tr* and *lb* *electromagnetic* deformons, lead to origination of **macroeffectons**, **macrotransitons** and **macrodeformons** (tr and lb respectively).

Correlated simultaneous excitations of tr and lb macroeffectons in the volume of superimposed *tr* and *lb* electromagnetic deformons lead to origination of **supereffectons**.

In turn, the coherent excitation of *both: tr and lb macrodeformons and macroconvertons* in the same volume means creation of **superdeformons**. Superdeformons are the biggest (cavitational) fluctuations, leading to microbubbles in liquids and to local defects in solids.

Total number of quasiparticles of condensed matter equal to $4!=24$, reflects all of possible combinations of the four basic ones [1-4], introduced above. This set of collective excitations in the form of "gas" of 3D standing waves of three types: de Broglie, acoustic and electromagnetic - is shown to be able to explain virtually all the properties of all condensed matter.

The important positive feature of our hierarchic model of matter is that it does not need the semi-empiric intermolecular potentials for calculations, which are unavoidable in existing theories of many body systems. The potential energy of intermolecular interaction is involved indirectly in dimensions and stability of quasiparticles, introduced in our model.

The main formulae of theory are the same for liquids and solids and include following experimental parameters, which take into account their different properties:

- [1]- **Positions of (tr) and (lb) bands in oscillatory spectra;**
- [2]- **Sound velocity;**
- [3]- **Density;**
- [4]- **Refraction index (extrapolated to the infinitive wave length of photon).**

The knowledge of these four basic parameters at the same temperature and pressure makes it possible using our computer program, to evaluate more than 300 important characteristics of any condensed matter. Among them are such as: total internal energy, kinetic and potential energies, heat-capacity and thermal conductivity, surface tension, vapor pressure, viscosity, coefficient of self-diffusion, osmotic pressure, solvent activity, etc. Most of calculated parameters are hidden, i.e. inaccessible to direct experimental measurement.

The new interpretation and evaluation of Brillouin light scattering and Mössbauer effect parameters may also be done on the basis of hierarchic theory. Mesoscopic scenarios of turbulence, superconductivity and superfluidity are elaborated.

Some original aspects of water in organization and large-scale dynamics of biosystems - such as proteins, DNA, microtubules, membranes and regulative role of water in cytoplasm, cancer development, quantum neurodynamics, etc. have been analyzed in the framework of Hierarchic theory.

Computerized verification of our Hierarchic theory of matter on examples of water and ice is performed, using special computer program: Comprehensive Analyzer of Matter Properties (CAMP, copyright, 1997, Kaivarainen). The new optoacoustic device (CAMP), based on this program, with possibilities much wider, than that of IR, Raman and Brillouin spectrometers, has been proposed (see URL: <http://www.karelia.ru/~alexk>).

This is the first theory able to predict all known experimental temperature anomalies for water and ice. The conformity between theory and experiment is very good even without any adjustable parameters.

The hierarchic concept creates a bridge between micro- and macro-phenomena, dynamics and thermodynamics, liquids and solids in terms of quantum physics.

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1. Theoretical background for macroscopic oscillations in condensed matter

One of the consequences of our concept is of special interest. It is the possibility for oscillation processes in solids and liquids. The law of energy conservation is not violated thereupon because the energies of two quasiparticle subsystems related to effectons and deformons, can change in opposite phases. The total internal energy of matter keeps almost constant.

The equilibrium shift between subsystems of condensed matter can be induced by any external factor, i.e. pressure or field. The relaxation time, necessary for system to restore its equilibrium, corresponding to minimum of potential or free energy after switching off external factor can be termed "memory" of system.

The energy redistribution between primary and secondary effecton and deformon subsystems may have a periodical character, coupled with the oscillation of the $(a \Leftrightarrow b)$ equilibrium constant of primary effectons ($K_{a \Leftrightarrow b}$) and correlated oscillations of primary electromagnetic deformons concentration if dissipation processes are weak or reversible. According to our model (Table 1 of [1, 2]) the $(a \rightarrow b)$ transition of primary effecton is related to photon absorption, i.e. a decrease in primary electromagnetic deformon concentration, while the $(b \rightarrow a)$ transition on the contrary, radiate photons. If, therefore, the $[a \Leftrightarrow b]$ and $[\bar{a} \Leftrightarrow \bar{b}]$ equilibriums are shifted right ward, and equilibrium constants $K_{a \Leftrightarrow b}$ and $\bar{K}_{a \Leftrightarrow b}$ decreases, then concentrations of primary and secondary deformons (n_d and \bar{n}_d) also decreases. If $K_{a \Leftrightarrow b}$ grows up, i.e. the concentration of primary effectons in a-states increases, then n_d increases. We remind that (a) and (b) states of

the primary effectons correspond to the more and less stable molecular clusters (see Introduction). In accordance with our model, the strong interrelation exists between dynamic equilibrium of primary and secondary effectons. Equilibrium of primary effectons is more sensitive to any perturbations. However, the equilibrium shift of secondary effectons affect the total internal energy, the entropy change and possible mass defect (see below) stronger than that of primary effectons.

As we have shown (Fig. 28a,b of [1] and [3]), the scattering ability of A-states is more than two times as high as that of B-states. Their polarizability, refraction index and dielectric permeability are also higher. It makes possible to register the oscillations in the condensed matter in different ways.

In accordance with our theory the oscillation of refraction index must induce the corresponding changes of viscosity and self-diffusion in condensed matter (see Chapter 11 of [1] and [4]). The diffusion variations are possible, for example, in solutions of macromolecules or other Brownian particles. In such a way self-organization in space and time gradually may originate in appropriate solvents, solutions, colloid systems and even in solid bodies.

The period and amplitude of these oscillations depend on the times of relaxation processes which are related to the activation energy of equilibrium shifts in the effectons, polyeffectons or coherent superclusters of primary effectons subsystems.

The reorganizations in the subsystems of translational and librational effectons, macro- and supereffectons, as well as chain-like polyeffectons, whose stabilities and sizes differ from each other, must go on at different rates. It should, therefore, be expected that in the experiment the presence of several oscillation processes would be revealed. These processes are interrelated but going with different periods and amplitudes. Concomitant oscillations of self-diffusion rate also must be taken into account. In such a way Prigogine's dissipative structures could be developed (Prigogine, 1984). Instability in the degree of ordering in time and space is accompanied by the slow oscillation of entropy of the whole macroscopic system.

The coherent extraterrestrial cosmic factors and gravitational instabilities can induce long relaxation and oscillation processes in water and other kind of condensed matter (Udaltsova, et. al., 1987).

2. The hypothesis of [entropy - mass - time] interrelation

The second law of thermodynamics for the closed system having the permanent number of particles and the constant internal energy is:

$$\frac{dQ_i}{T} = dS_i \geq 0 \quad (1)$$

where: dQ_i appears to be due to the irreversible processes within the system and is referred to as *uncompensated heat* (Prigogine, 1980, 1984, Babloyantz, 1986).

This law means the only possibility of the processes, accompanied by the increase of entropy (S) (related neither to chemical or nuclear reactions).

The statistical interpretation of entropy is expressed with the Boltzmann formula:

$$S = k \cdot \ln P \simeq k \cdot \ln W \quad (2)$$

where: k is the Boltzmann constant, P is the statistical weight, which is proportional to the number of realizations (number of microstates) for the given state of the macroscopic system (W).

Correspondingly the probability of one of this microstates is: $p_i = 1/W$.

It follows from (2) that the second law of thermodynamics expresses the fact that the system tends to the most probable state.

$$dS = k \, d \ln W \geq 0 \quad (3)$$

or

$$dQ_i = T dS_i = kT \, d \ln W_i \quad (4)$$

Boltzmann has put forward the hypothesis that the irreversibility (i.e. asymmetry) of time is determined with the irreversibility of processes according to the second law of thermodynamics. Prigogine has modified and developed this idea (Prigogine, 1984), introducing the notions of the **internal time and microscopic operator of entropy**. The "ARROW OF TIME" and irreversibility is a result of asymmetry of physically accessible states after Prigogin. The microscopic mechanism of irreversibility is discussed also in Chapter 8 of Part II of book [1].

It follows from the second law and formulae (1.12) from Part II of this book, that the mass of a system of N similar particles with the mass ($M \simeq Nm$) either does not change in the course of time, or decreases:

$$\frac{dt}{t} = -\frac{1}{2} \frac{dM}{M} \geq 0, \quad (5)$$

where: dM is the change in the mass of a macroscopic system related to electromagnetic and acoustic radiation, and also to intermolecular interaction induced small mass defect.

It follows from (5) that the positive pace of time in the system is related to the decrease in its mass, while the negative pace of time - to the increase in its mass. Hence, the coherent oscillation processes in the system (which

are not related to chemical reactions) are related to oscillation of mass [M]. It is known that phonons and photons do not have the rest masses. Therefore, the coherent periodical energy exchange between subsystems of effectons and deformons (primary and secondary) must be accompanied by changes in the mass of the effecton subsystem and total system.

The macroscopic fluctuations of mass values for solid bodies were registered experimentally indeed (Kozyrev, 1958).

The oscillations of temperature and thermal conductivity also must accompany the oscillation of secondary acoustic deformons concentration.

The total internal energy of the closed system can be represented as a sum of contributions from the primary (U^a and U^b), secondary (\bar{U}^a and \bar{U}^b), macro (U^A, U^B) and super (U^{A*}, U^{B*}) effectons, convertons and contributions of corresponding deformons and transitons (see formula 4.3).

At $U_{\text{tot}} = \text{const}$ the exchange of energies between the subsystems of effectons and deformons can be approximately represented as:

$$-d(mc_x^2)N \approx d(h\bar{\nu}_d)N \quad (6)$$

where: N is the number of particles in the system;

$[c_x = (v_{gr}v_{ph})^{1/2} = \text{const}]$ is the characteristic wave B velocity in the system, which is equal to the product of the generalized group (v_{gr}) and phase (v_{ph}) velocities of wave B of particles; $[d(h\bar{\nu}_d)N]$ is the change of energy contribution of secondary deformons, which is much bigger than that of primary deformons due to low concentration of latter; $[dm]$ is the defect of mass per particle in the system as a result of energy exchange between subsystems of the effectons and deformons.

Because phonons are carriers of the heat energy, then the uncompensated heat (dQ_i) in eq.(1) could be easily related to the irreversible increment of secondary acoustic deformons and convertons energy or to the uncertainty in this energy:

$$dQ_i = TdS = Nd(h\bar{\nu}_d)_{tr,lb} \quad (7)$$

or

$$dQ_i = TdS = -d(mc_x^2)N = -d(Mc_x^2)_{tr,lb} \quad (8)$$

$$\text{where: } \bar{\nu}_d = [(\bar{\nu}_d)_{tr,lb} + (\bar{\nu}_d)_{ca,cb,cMd}] + (\nu_d)_{tr,lb}$$

$(\bar{\nu}_d)_{tr,lb}$ is the resulting frequency of secondary deformons (tr and lb);
 $(\bar{\nu}_d)_{ca,cb,cMd}$ is the resulting frequency of convertons and related deformons;
 $M = Nm$ is the total mass of particles in the system.

$(\nu_d)_{tr,lb}$ are a frequencies of tr and lb IR photons.

Let us enter the notion of absolute entropy (S) as a measure of the uncompensated heat change $(\Delta Q_i/T)$, which could occur at $(\bar{a} \Leftrightarrow \bar{b})$ transitions of mean effectons and convertons subsystems at the given temperature:

$$S = \int \frac{dQ_i}{T} \quad (9)$$

Putting (7) and (8) into (9) and multiplying numerator and denominator, to Boltzmann constant (k) keeping in mind (5), we derive a new approximate formula for absolute entropy:

$$S = \frac{\Delta Q}{T} = \frac{N \Delta (3h\bar{\nu}_d)}{T} \approx 3Nk \left(\frac{h\bar{\nu}^a}{kT} \right) \approx -\frac{\Delta M c_x^2}{T} = \frac{2M}{T} \frac{\Delta t}{t_{tr,lb}} \quad (10)$$

where: $\bar{\nu}_d = |\bar{\nu}_a - \bar{\nu}_b| \sim \bar{\nu}_a$ is a frequency of secondary acoustic deformon; $\bar{\nu}_a$ is calculated from formula (2.54).

Formula (10) relates the positive entropy value not only to heat radiation from the system or the increase of the deformons contribution into the total internal energy ($\Delta Q_i > 0$), determined the corresponding decrease of its mass ($\Delta M < 0$), but also to positive time course in this system ($\Delta t > 0$).

The interesting experiments done by Kosyrev (1958) confirm one of the consequences of our theory, that the shift of equilibrium between subsystems of the effectons and acoustic deformons to the latter ones should decrease the mass of body. In his experiments with special balance it was shown that the activation of phonons with sufficiently high energy by means of sound generator - decreases the mass of solid body: (100 – 800) g. The value of decreasing was proportional to the total mass of body. The relative mass decreasing was estimated as:

$$\Delta M/M = (2.3 - 3.4) \cdot 10^{-5}$$

In liquids and solids, the concentration of secondary deformons can be lower than the concentration of molecules. Therefore, in a general case N in the formula (10) must be substituted by the number of secondary deformons in the system:

$$(\bar{N}_d)_{tr,lb} = V(\bar{n}_d)_{tr,lb}, \quad (11)$$

where V is the volume of the system;

$$(\bar{n}_d)_{tr,lb} = \frac{8}{9} \pi \left(\frac{\bar{\nu}_d}{v_{res}} \right)_{tr,lb}^3$$

is the concentration of secondary translational and librational effectons;

$$\nu_{ph}^{res} = (\bar{\nu}_{ph}^1 \bar{\nu}_{ph}^2 \bar{\nu}_{ph}^3)^{1/3} \quad (12)$$

is the resulting frequency of secondary deformons; $\bar{\nu}_{ph}^{1,2,3}$ are calculated from (3.15); ν_{res} is the resulting thermal phonons velocity, which is equal to isotropic hypersonic velocity in liquids and to transversal one (v) in solids.

The total entropy of a condensed substance (S_{tot}) is approximately a sum of secondary effecton contribution (*tr and lb*) and contribution of convertions. Using (10) and (11), we obtain:

$$\begin{aligned} S_{tot} \approx S_{tr} + S_{lb} + S_{con} = \frac{V_0 h}{T} [\bar{n}_d^{tr} (3 \bar{\nu}_d^{res})_{tr} + \bar{n}_d^{lb} (3 \bar{\nu}_d^{res})_{lb}] + \\ + \frac{V_0 h}{T} n_{con} 3 [(\nu_{ef}^a)_{tr} - (\nu_{ef}^a)_{lb}]_{con} \end{aligned} \quad (13)$$

where: \bar{n}_d^{tr} and \bar{n}_d^{lb} correspond to (11); n_{con} is a concentration of convertions, equal to that of primary librational effectons (n_{lb});

$(\bar{\nu}_d)_{tr,lb}^{res}$ are the resulting frequencies of secondary deformons (tr and lb).

Knowing the positions of translational and librational bands in the oscillatory spectra and sound velocity, one can estimate the entropy of matter at each temperature using (13).

At constant pressure and volume the change of enthalpy (H) is equal to the change of internal energy (U). Therefore, the formulae that we have obtained for (U) and (S) allow to calculate also changes in free energy:

$$\Delta G_{P,V} = \Delta U - T \Delta S \quad (14)$$

The qualitative correctness of the formula obtained for entropy is obvious from (10) in the form:

$$S_{tr,lb} \approx Nk(3h\bar{\nu}^a/kT)_{tr,lb} + Nk(3h\Delta\nu^a/kT)_{acon} \simeq Nk(3h\nu^a/kT)_{tr,lb} \quad (15)$$

It follows from (2.54) and (15) that:

- a) at $T \rightarrow 0$: $h\bar{\nu}^a/kT \rightarrow 0$ and $S \rightarrow 0$;
- b) at decreasing ν_p in the melting point at the growth of T both $h\bar{\nu}_a/kT$ and S grow up;
- c) if the mixing of liquids and gases leads to the weakening of pair interaction in effectons or clusters, then $\nu_p = (E_b - E_a)/h$ decreases and S increases.

Equalizing (2) and right part of (15) we have in the framework of our approximations:

$$S = k \ln P \approx k \left(N_0 \frac{h\bar{\nu}_a}{kT} \right) = R \frac{h\bar{\nu}_a}{kT} \quad (16)$$

from (16) we can derive an approximate formula for statistical weight:

$$P \approx \exp \left(N_0 \frac{h\bar{\nu}_a}{kT} \right)_{tr,lb} \quad (17)$$

According to (16) the law of entropy growth *means the striving of the real quantum properties of the substance to ideal properties*. It means the tending of primary effectons energy in the (a) state ($h\nu_a$) to the thermal equilibrium value (kT):

$$\Delta S > 0, \quad \text{if} \quad [h\bar{\nu}^a \rightarrow kT] \quad (18)$$

Competition between the discrete quantum energy distribution and its tendency to kT may be a reason for instability of different parameters of condensed matter. This may lead to origination of macroscopic oscillations in the system interrelated with entropy, temperature and mass (eq.10). Such oscillations could be considered as a kind of self-organization process due to feedback links in a hierarchic system of interrelated quasiparticles of matter.

3. The entropy - information content of matter as a hierarchic system

The statistical weigh for macrosystem (P), equal to number of microstates (W), corresponding to given macrostate, necessary for entropy calculation using (2) could be presented as:

$$W = \frac{N!}{N_1! \cdot N_2! \cdot \dots \cdot N_q!} \quad (19)$$

where:

$$N = N_1 + N_2 + \dots N_q \quad (20)$$

is the total number of molecules in macrosystem;

N_i is the number of molecules in the i-th state;

q is the number of independent states of all quasiparticles in macrosystem.

We can subdivide macroscopic volume of 1cm^3 into 24 types of quasiparticles in accordance with our hierarchic model (see Table 1 of [1, 2]).

In turn, each type of the effectons (primary, secondary, macro- and super-effectons) is subdivided on two states: ground (a,A) and excited (b,B) states. Taking into account two ways of the effectons origination - due to thermal translations (tr) and librations (lb), excitations, related to $[lb/tr]$ convertions, macro- and super deformons, the total number of **independent** states is 24 also. It is equal to number of independent relative probabilities of excitations, composing partition function Z (see eq.4.2 of [1, 2]). Consequently, in eqs.(4.19 and 4.20) we have:

$$q = 24$$

The **number** of molecules, in the unit of volume of condensed matter (1cm^3), participating in each of 24 excitation states (i) can be calculated as:

$$N_i = \frac{(v)_i}{V_0/N_0} \cdot n_i \cdot \frac{P_i}{Z} = \frac{N_0}{V_0} \frac{P_i}{Z} \quad (21)$$

where: $(v)_i = 1/n_i$ is the volume of (i) quasiparticle, equal to reciprocal value of its concentration (n_i) ; N_0 and V_0 are Avogadro number and molar volume, correspondingly; Z is partition function and P_i are relative probabilities of independent excitations in composition of Z (eq.4.2).

The total number of molecules of (i)-type of excitation in any big volume of matter (V_{Mac}) is equal to

$$N_{\text{Mac}}^i = N_i V_{\text{Mac}} = V_{\text{Mac}} \frac{N_0}{V_0} \frac{P_i}{Z} \quad (21a)$$

Putting (20) into (18) and (19), we can calculate the statistical weight and entropy from eq.(2).

For large values of N_i it is convenient to use a Stirling formula:

$$N_i = (2\pi N)^{1/2} (N/e)^N \cdot \exp(\Theta/12N) \sim (2\pi N)^{1/2} (N/\Theta)^N \quad (21b)$$

Using this formula and (20), one can obtain the following expression for entropy:

$$S = k \cdot \ln W = -k \cdot \sum_i^q (N_i + \frac{1}{2}) \ln N_i + \text{const} = S_1 + S_2 + \dots S_i \quad (22)$$

From this eq. we can see that the temperature increasing or [solid \rightarrow liquid] phase transition will lead to the entropy elevation:

$$\Delta S = S_L - S_S = k \cdot \ln(W_L/W_S) > 0 \quad (23)$$

It follows from (22, 20) and (19) that under conditions when (P_i) and N_i undergoes oscillations it can lead to oscillations of contributions of different types of quasiparticles to the entropy of system and even to oscillations of total entropy of system as an additive parameter. The coherent oscillations of P_i and N_i can be induced by different external fields: acoustic, electromagnetic and gravitational. Macroscopic autooscillations may arise spontaneously also in the sensitive and highly cooperative systems.

Experimental evidence for such phenomena will be discussed in the next section.

The notions of probability of given microstate ($p_i = 1/W$), entropy (S_i) and information (I_i) are strongly interrelated. The smaller the probability the greater is information (Nicolis 1986):

$$I_i = \lg_2 \frac{1}{p_i} = -\lg_2 p_i = \lg_2 W_i \quad (24)$$

where p_i is defined from the Boltzmann distribution as:

$$p_i = \frac{\exp(-E_i/kT)}{\sum_{m=0}^{\infty} \exp(-n_m h\nu_i/kT)} \quad (25)$$

where n_m is quantum number; h is the Plank constant; $E_i = h\nu_i$ is the energy of (i)-state.

There is strict relation between the entropy and information, leading from comparison of (24) and (2):

$$S_i = (k_B \ln 2) I_i = 2.3 \cdot 10^{-24} I_i \quad (26)$$

The information entropy is given as expectation of the information in the system (Nicolis, 1986; Haken, 1988).

$$\langle I \rangle = \sum P_i \lg_2(1/p_i) = -\sum p_i \lg_2(p_i) \quad (27)$$

From (26) and (22) we can see that variation of probability p_i and/or N_i in (20) will lead to changes of entropy and information, characterizing the matter as a hierarchical system.

The **reduced information** (entropy), characterizing its **quality**, related to selected collective excitation of any type of condensed matter, we introduce here as a product of corresponding component of information $[I_i]$ to the number of molecules (atoms) with similar dynamic properties in composition of this excitation:

$$q_i = (v_i/v_m) = N_0/(V_0 n_i) \quad (27a)$$

where: $v_i = 1/n_i$ is the volume of quasiparticle, reversible to its concentration (n_i); $v_m = V_0/N_0$ is the volume, occupied by one molecule.

The product of (27) and (27a), i.e. the **reduced information** gives the quantitative characteristic not only about quantity but also about the quality of the information:

$$(Iq)_i = p_i \lg_2(1/p_i) \cdot N_0/(V_0 n_i) \quad (27b)$$

This new formula could be considered as a useful modification of known Shannon equation.

4. Experimentally revealed macroscopic oscillations

A series of experiments was conducted in our laboratory to study oscillations in the buffer (pH 7.3) containing 0.15 M NaCl as a control system and immunoglobulin G solutions in this buffer at the following concentrations: $3 \cdot 10^{-3}$; $6 \cdot 10^{-3}$; $1.2 \cdot 10^{-2}$ and $2.4 \cdot 10^{-2} mg/ml$.

The turbidity (D^*) of water and the solutions were measured every 10 seconds with the spectrophotometer at $\lambda = 350 nm$. Data were obtained automatically with the time constant 5 s during 40 minutes. The number of D^* values in every series was usually equal to 256. The total number of the fulfilled series was more than 30.

The time series of D^* were processed by the software for time series analysis. The time trend was thus subtracted and the autocovariance function and the spectral density were calculated.

The empty quartz cuvette with the optical path about 1 cm were used as a basic control.

Only the optical density of water and water dissolved substances, which really exceeded background optical density in the control series were taken into account. It is shown that the noise of the photoelectronic multiplier does not contribute markedly to dispersion of D^* .

The measurements were made at temperatures of 17, 28, 32, and 37°. The period of the trustworthily registered oscillation processes related to changes in D^* , had 2 to 4 discrete values over the range of (30 – 600) s under our conditions. It does not exclude the fact that the autooscillations of longer or shorter periods exist. For example, in distilled water at 32°C the oscillations of the scattering ability are characterized by periods of 30, 120 and 600 s and the spectral density amplitudes 14, 38 and 78 (in relative units), respectively. With an increase in the oscillation period their amplitude also increases. At 28°C the periods of the values 30, 41 and 92s see have the corresponding normalized amplitudes 14.7, 10.6 and 12.0.

Autooscillations in the buffer solution at 28°C in a 1 cm wide cuvette with the optical way length 1 cm (i.e. square section) are characterized with periods: 34, 52, 110 and 240 s and the amplitudes: 24, 33, 27 and 33 relative units. In the cuvette with a smaller (0.5 cm) or larger (5 cm) optical wavelength at the

same width (1 cm) the periods of oscillations in the buffer change insignificantly. However, amplitudes decreased by 50% in the 5 cm cuvette and by 10-20% in the 0.5 cm-cuvette. This points to the role of geometry of space where oscillations occur, and to the existence of the finite correlation radius of the synchronous processes in the volume. But this radius is macroscopic and comparable with the size of the cuvette.

The dependence of the autooscillations amplitude on the concentration of the protein - immunoglobulin G has a sharp maximum at the concentration of $1.2 \cdot 10^{-2} \text{ mg/ml}$. There is a background for considering it to be a manifestation of the hydrodynamic Bjorkness forces between the pulsing macromolecules (Käiväräinen, 1987).

Oscillations in water and water solutions with nearly the same periods have been registered by the light-scattering method by Chernikov (1985).

Chernikov (1990d) has studied the dependence of light scattering fluctuations on temperature, mechanic perturbation and magnetic field in water and water hemoglobin and DNA solution. It has been shown that an increase in temperature results in the decline of long-term oscillation amplitude and in the increase of short-time fluctuation amplitude. Mechanical mixing removes long-term fluctuations and over 10 hours are spent for their recovery. Regular fluctuations (oscillations) appear when the constant magnetic field above 240 A/m is applied; the fluctuations are retained for many hours after removing the field. The period of long-term oscillations has the order of 10 minutes. It has been assumed that the maintenance of long-range correlation of molecular rotation-translation fluctuation underlies the mechanism of long-term light scattering fluctuations.

It has been shown (Chernikov, 1990b) that a pulsed magnetic field (MF), like constant MF, gives rise to light scattering oscillations in water and other liquids containing H atoms: glycerin, xylol, ethanol, a mixture of unsaturated lipids. All this liquids also have a distinct response to the constant MF. "Spontaneous" and MF-induced fluctuations are shown to be associated with the isotropic component of scattering. These phenomena do not occur in the nonproton liquid (carbon tetrachloride) and are present to a certain extent in chloroform (containing one hydrogen atom in its molecule). The facts obtained indicate an important role of hydrogen atoms and cooperative system of hydrogen bonds in "spontaneous" and induced by external perturbations macroscopic oscillations.

The understanding of such phenomena can provide a physical basis for of self-organization (Prigogine, 1980, 1984, Babloyantz, 1986), the biological system evolution (Shnol, 1979, Udaltsova et al., 1987), and chemical processes oscillations (Field and Burger, 1988).

It is quite probable that macroscopic oscillation processes in biological liquids, e.g. blood and liquor, caused by the properties of water are involved in animal and human physiological processes.

We have registered the oscillations of water activity in the protein-cell system by means of light microscopy using the apparatus "Morphoquant", through the change of the erythrocyte sizes, the erythrocytes being ATP-exhausted and fulfilling a role of the passive osmotic units. The revealed oscillations have a

few minute-order periods.

Preliminary data obtained from the analysis of oscillation processes in the human cerebrospinal liquor indicate their dependence on some pathology. Perhaps, the autooscillations spectrum of the liquor can serve as a sensitive test for the physiological status of the organism. The liquor is an electrolyte and its autooscillations can be modulated with the electromagnetic activity of the brain.

We suggested that the activity of the central nervous system and the biological rhythms of the organism are dependent with the oscillation processes in the liquor. If it is the case, then the directed influence on these autooscillation processes, for example, by means of magnetic field makes it possible to regulate the state of the organism and its separate organs. Some of reflexotherapeutic effects can be caused by correction of biorhythms.

During my stay in laboratory of Dr. G.Salvetty in the Institute of Atomic and Molecular Physics in Pisa (Italy) in 1992, the oscillations of heat capacity $[C_p]$ in 0.1 M phosphate buffer (pH7) and in 1% solution of lysozyme in the same buffer at $20^{\circ}C$ were revealed. The sensitive adiabatic differential microcalorimeter was used for this aim. The biggest relative amplitude changing: $[\Delta C_p]/[C_p] \sim (0.5 \pm 0.02)\%$ occurs with period of about 24 hours, i.e. corresponds to circadian rhythm.

Such oscillations could be stimulated by the variation of magnetic and gravitational conditions of the Earth during this period.

5. Phenomena in water and aqueous systems, induced by magnetic field

In the works of (Semikhina and Kiselev, 1988, Kiselev et al., 1988, Berezin et al., 1988) the influence of the weak magnetic field was revealed on the dielectric losses, the changes of dissociation constant, density, refraction index, light scattering and electroconductivity, the coefficient of heat transition, the depth of super-cooling for distilled water and for ice also. This field used as a modulator a geomagnetic action.

The absorption and the fluorescence of the dye (rhodamine 6G) and protein in solutions also changed under the action of weak fields on water. The latter circumstance reflects feedback links in the guest-host, or solute -solvent system.

The influence of constant and variable magnetic fields on water and ice in the frequency range $10^4 - 10^8 Hz$ was studied. The maximum sensitivity to field action was observed at the frequency $\nu_{\max} = 10^5 Hz$. In accordance with our calculations, this frequency corresponds to frequency of superdeformons excitations in water (see Fig. 48d of [1] and article [2]).

A few of physical parameters changed after the long (nearly 6 hour) influence of the variable fields (\tilde{H}), modulating the geomagnetic field of the tension $[H = H_{\text{geo}}]$ with the frequency (f) in the range of $(1 - 10) \cdot 10^2 Hz$ (Semikhina and Kiselev, 1988, Kiselev et al., 1988):

$$H = H \cos 2\pi ft \quad (28)$$

In the range of modulating magnetic field (H) tension from 0.08 A/m to 212A/m the **eight maxima of dielectric losses tangent** in the above mentioned (f) range were observed. Dissociation constant decreases more than other parameters (by 6 times) after the incubation of ice and water in magnetic field. The relaxation time ("memory") of the changes, induced in water by fields was in the interval from 0.5 to 8 hours.

The authors interpret the experimental data obtained as the influence of magnetic field on the probability of proton transfer along the net of hydrogen bonds in water and ice, which lead to the deformation of this net.

The *equilibrium constant* for the reaction of dissociation:



in ice is less by almost six orders ($\simeq 10^6$) than that for water. On the other hand the values of the *field- induced effects in ice are several times more than in water*, and the time for reaching them in ice is less. So, the above interpretation is doubtful.

In the framework of our concept all the aforementioned phenomena could be explained by the shift of the ($a \rightleftharpoons b$) equilibrium of primary translational and librational effectons to the left. In turn, this shift stimulates polyeffectons or coherent superclusters growth, under the influence of magnetic fields. Therefore, parameters such as the refraction index, dielectric permeability and light scattering have to enhance symbatically, while the H_2O dissociation constant depending on the probability of superdeformons must decrease. The latter correlate with declined electric conductance.

As far, the magnetic moments of molecules within the coherent superclusters or polyeffectons formed by primary librational effectons are additive, then the values of changes induced by magnetic field must be proportional to polyeffecton sizes. These sizes are markedly higher in ice than in water and decrease with increasing temperature.

Inasmuch the effectons and polyeffectons interact with each other by means of phonons (i.e. the subsystem of secondary deformons), and the velocity of phonons is higher in ice than in water, then the saturation of all concomitant effects and achievement of new equilibrium state in ice is faster than in water.

The frequencies of geomagnetic field modulation, at which changes in the properties of water and ice have maxima can correspond to the eigen-frequencies of the [$a \rightleftharpoons b$] equilibrium constant of primary effectons oscillations, determined by [assembly \rightleftharpoons disassembly] equilibrium oscillations for coherent super clusters or polyeffectons.

The presence of dissolved molecules (ions, proteins) in water or ice can influence on the initial [$a \rightleftharpoons b$] equilibrium dimensions of polyeffectons and, consequently the interaction of solution with outer field.

Narrowing of ^1H -NMR lines in a salt-containing water and calcium bicarbonate solution was observed after magnetic field action. This indicates that the degree of ion hydration is decreased by magnetic treatment. On the other hand, the width of the resonance line in *distilled water* remains unchanged after 30 minute treatment in the field (135 kA/m) at water flow rate of 60 cm/s (Klassen, 1982).

The hydration of diamagnetic ions (Li^+ , Mg^{2+} , Ca^{2+}) decreases, while the hydration of paramagnetic ions (Fe^{3+} , Ni^{2+} , Cu^{2+}) increases. It leads from corresponding changes in ultrasound velocity in ion solutions (Duhanin and Kluchnikov, 1975).

There are numerous data which pointing to an increase the coagulation of different particles and their sedimentation velocity after magnetic field treatment. These phenomena provide a reducing the scale formation in heating systems, widely used in practice. Crystallization and polymerization also increase in magnetic field. It points to decrease of water activity.

Increasing of refraction index (n) and dielectric permeability ($\epsilon \simeq n^2$) and symbatic enhancement of water viscosity (Minenko, 1981) are in total accordance with our viscosity theory (eqs. 11.44 and 11.45 of [1] and article [4]).

It follows from our mesoscopic model that the increase of (n) is related to the increase of molecular polarizability (α) due to the shift of $(a \rightleftharpoons b)_{tr,lb}$ equilibrium of primary effectons leftward under the action of magnetic field. On the other hand, distant Van der Waals interactions and consequently dimensions of primary effectons depend on α . This explains the elevation of surface tension of liquids after magnetic treatment (see Chapter 11 of [1] or [4]).

The leftward shift of $(a \rightleftharpoons b)_{tr,lb}$ equilibrium of primary effectons must lead to decreasing of water activity due to (n^2) increasing and structural factor (T/U_{tot}) decreasing its structure ordering. Corresponding changes in the vapor pressure, freezing, and boiling points, coagulation, polymerization and crystallization are the consequences of this shift and water activity decreasing.

It follows from mesoscopic theory that any changes in condensed matter properties must be accompanied by change of such parameters as:

- 1) density;
- 2) sound velocity;
- 3) positions of translational and librational bands in oscillatory spectra;
- 4) refraction index.

Using our equations and computer simulations by means of elaborated software (CAMP: Comprehensive Analyzer of Matter Properties), it is possible to obtain from these changes very detailed information (more than 200 parameters) about even small perturbations of matter on meso- and macroscopic levels.

Available experimental data indicate that all of above mentioned 4 experimental parameters of water have been changed indeed after magnetic treatment.

Minenko (1981) has shown that bidistilled water *density* increases by about 0.02% after magnetic treatment (540 kA/m, flow rate 80 cm/s).

Sound velocity in distilled water increases to 0.1% after treatment under conditions: 160 kA/m and flow rate 60 cm/s.

The *positions of the translational and librational bands* of water were also changed after magnetic treatment in 415 kA/m (Klassen, 1982).

Coherent radio-frequency oscillations in water, revealed by C. Smith

It was shown experimentally by C. Smith (1994) that the water display a coherent properties. He shows that water is capable of retaining the frequency of an alternating magnetic field. For a tube of water placed inside a solenoid coil, the threshold for the alternating magnetic field, potentiating electromagnetic frequencies into water, is $7.6 \mu T$ (rms). He comes to conclusion that the frequency information is carried on the magnetic vector potential.

He revealed also that in a course of yeast cells culture synchronously dividing, the radio-frequency emission around 1 MHz (10^6 1/s), 7-9 MHz (7.9×10^6 1/s) and 50-80 MHz (5.9×10^7 1/s) with very narrow bandwidth (~ 50 Hz) might be observed for a few minutes.

These frequencies could correspond to frequencies of different water collective excitations, introduced in our Hierarchic theory, like [lb/tr] macroconvertons, the $[a \rightleftharpoons b]_{lb}$ transitons, etc. (see Fig. 48 of [1] and [2]), taking into account the deviation of water properties in the colloid and biological systems as respect to pure one.

Cyril Smith has proposed that the increasing of coherence radius in water could be a consequence of coherent water clusters association due to Josephson effect (Josephson, 1965): tunneling of molecules between clusters. As far primary librational effectons are resulted from partial Bose-condensation of molecules, this idea looks quite acceptable in the framework of our Hierarchic theory.

The coherent oscillations in tube with water, revealed by C. Smith could be induced by coherent electromagnetic radiation of microtubules of cells, produced by correlated intra-MTs water excitations (see Section 17.5 and Fig. 48 of [1]).

The biological effects of magnetically treated water are very important practically. For example, hemolysis of erythrocytes is more vigorous in magnetically pretreated physiological solutions (Trincher, 1967). Microwave radiation induces the same effect (Il'ina et al., 1979). But after boiling such effects in the treated solutions have been disappeared. It is shown that magnetic treatment of water strongly stimulates the growth of corn and plants (Klassen, 1982).

Now it is obvious that a systematic research program is needed to understand the physical background of multilateral effects of magnetized water.

6. Influence of weak magnetic field on the properties of solid bodies

It has been established that as a result of magnetic field action on solids with interaction energy ($\mu_B H$) much less than kT , many properties of matter such as hardness, parameters of crystal cells and others change significantly.

The short-time action of magnetic field on silicon semiconductors is followed by a very long (many days) relaxation process. The action of magnetic field was in the form of about 10 impulses with a length of 0.2 ms and an amplitude of about $10^5 A/m$. The most interesting fact was that this relaxation had an oscillatory character with periods of about several days (Maslovsky and Postnikov, 1989).

Such a type of long period oscillation effects has been found in magnetic and nonmagnetic materials.

This points to the general nature of the macroscopic oscillation phenomena in solids and liquids.

The period of oscillations in solids is much longer than in liquids. This may be due to stronger deviations of the energy of (a) and (b) states of primary effectons and polyeffectons from thermal equilibrium and much lesser probabilities of transition and deformation excitation. Consequently, the relaxation time of ($a \leftrightarrow b$)_{tr,lb} equilibrium shift in solids is much longer than in liquids. The oscillations originate due to instability of dynamic equilibrium between the subsystems of effectons and deformons.

7. Possible mechanism of perturbations of nonmagnetic materials under magnetic treatment

We shall try to discuss the interaction of magnetic field with diamagnetic matter like water as an example. The magnetic susceptibility (χ) of water is a sum of two opposite contributions (Eisenberg and Kauzmann, 1969):

1) average negative diamagnetic part, induced by external magnetic field:

$$\bar{\chi}^d = \frac{1}{2}(\chi_{xx} + \chi_{yy} + \chi_{zz}) \cong -14.6(\pm 1.9) \cdot 10^{-6}$$

2) positive paramagnetism related to the polarization of water molecule due to asymmetry of electron density distribution, existing without external magnetic field. Paramagnetic susceptibility (χ^p) of H_2O is a tensor with the following components:

$$\chi_{xx}^p = 2.46 \cdot 10^{-6}; \quad \chi_{yy}^p = 0.77 \cdot 10^{-6}; \quad \chi_{zz}^p = 1.42 \cdot 10^{-6} \quad (29)$$

The resulting susceptibility:

$$\chi_{H_2} = \bar{\chi}^d + \bar{\chi}^p \cong -13 \cdot 10^{-6}$$

The second contribution in the magnetic susceptibility of water is about 10 times lesser than the first one. But the first contribution to the magnetic moment of water depends on external magnetic field and must disappear when it is switched out in contrast to second one.

The coherent primary librational effectons of water even in liquid state contain about 100 molecules $[(n_M^{ef})_{lb} \simeq 100]$ at room temperature (Fig. 7a of [1] or Fig.4a of [2]). In ice $(n_M^{ef})_{lb} \geq 10^4$. In (a) -state the vibrations of all these molecules are synchronized in the same phase, and in (b) -state - in counterphase. Correlation of H_2O forming effectons means that the energies of interaction of water molecules with external magnetic field are additive:

$$\epsilon^{ef} = n_M^{ef} \cdot \mu_p H \quad (30)$$

In such a case this total energy of effecton interaction with field may exceed thermal energy:

$$\epsilon^{ef} > kT \quad (31)$$

In the case of polyeffectons formation this inequality becomes much stronger.

It follows from our model that interaction of magnetic field with (a) -state of the effectons must be stronger than that with (b) -state due to the additivity of the magnetic moments of coherent molecules:

$$\epsilon_a^{ef} > \epsilon_b^{ef} \quad (32)$$

Consequently, magnetic field shifts $(a \Leftrightarrow b)_{tr,lb}$ equilibrium of the effectons leftward. At the same time it minimizes the potential energy of matter, because potential energy of (a) -state (V_a) is lesser than (V_b):

$$V_a < V_b \quad \text{and} \quad E_a < E_b, \quad (33)$$

where $E_a = V_a + T_{\text{kin}}^a$; $E_b = V_b + T_{\text{kin}}^b$ are total energies of the effectons.

We keep in mind that the kinetic energies of (a) and (b) -states are equal: $T_{\text{kin}}^a = T_{\text{kin}}^b = p^2/2m$.

These energies decreases with increasing of the effectons dimensions, determined by the most probable impulses in selected directions:

$$\lambda_{1,2,3} = h/p_{1,2,3}$$

The energy of interaction of magnetic field with deformons as a transition state of effectons must be even less than ϵ_b^{ef} due to lesser order of molecules in this state and reciprocal compensation of their magnetic moments:

$$\epsilon_d < \epsilon_b^{ef} \leq \epsilon_a^{ef} \quad (34)$$

This important inequality means that as a result of external magnetic field action the shift of $(a \leftrightarrow b)_{tr,lb}$ leftward is reinforced by leftward shift of equilibrium [effectons \rightleftharpoons deformons] subsystems of matter.

If water is flowing in a tube it increases the relative orientations of all effectons in volume and stimulate the coherent superclusters formation. All the above discussed effects must increase. Similar ordering phenomena happen in a rotating tube with liquid.

After switching off the external magnetic field the relaxation of *induced ferromagnetism* in water begins. It may be accompanied by the oscillatory behavior of $(a \leftrightarrow b)_{tr,lb}$ equilibrium. All the experimental effects discussed above can be explained as a consequence of orchestrated in volume $(a \leftrightarrow b)$ equilibrium oscillations.

Remnant ferromagnetism in water was experimentally established using a SQUID superconducting magnetometer by Kaivarainen et al. in 1992 (unpublished data). Water was treated in constant magnetic field 50G for two hours. Then it was frozen and after switching off external magnetic field the remnant ferromagnetism was registered at helium temperature. Even at this low temperature a slow relaxation time- dependent decrease of ferromagnetic signal was revealed. These results point to the correctness of the proposed mechanism of magnetic field - water interaction.

The attempt to make a theory of magnetic field influence on water, based on other model were made earlier (Yashkichev, 1980). However, this theory does not take into account the quantum properties of water and cannot be considered as satisfactory one.

The comprehensive material obtained by Udaltsova, Kolombet and Shnol (1987) when studying various macroscopic oscillations reveals their fundamental character and their dependence on gravitation factor.

The correlated changes of time, entropy and mass of any condensed matter follows from our theory.

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